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FINAL PERFORMANCE REPORT

Grant AFOSR FA9550-07-1-0103

Physics of Spin-Polarized Media

William Happer

Department of Physics

Princeton University

March 6, 2011

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ANNUAL PERFORMANCE REPORT

Grant AFOSR FA9550-07-1-0103

Physics of Spin-Polarized Media

by

William Happer
Department of Physics
Princeton University

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Overview

The work supported by this AFOSR grant during the past year was focused on the fundamental physics and the applications of spin polarized species. The most notable accomplishments of the past year were: (1) basic work basic physics needed to transform gas-cell atomic clocks based on optically-pumped alkali-metal atoms from secondary frequency standards to primary frequency standards; (2) completion of studies of optical pumping and magnetic resonances of spin-polarized metastable ^{129}Xe atoms as potential candidates for atomic clocks; (3) detailed studies of nuclear and electron spin currents from an optically pumped vapor to cell walls; (4) initial studies of the optical pumping of alkali-metal atoms under laboratory conditions similar to those prevailing in sodium guidestars used in adaptive optics; (5) Personnel supported by this AFOSR grant or its predecessors have published a book “Optical Pumping of Atoms,” based on work that AFOSR has supported over the years. We will provide a brief overview of these main areas below, and we will provide citations where more details can be found from papers we have published.

Most of the work supported by this AFOSR grant has had and will continue to have applications to technologies of importance to the Department of Defense and to the United States Air Force. For example, atomic clocks based on optically-pumped alkali-metal atoms are by far the most widely used atomic clocks for applications in support of US Air Force missions. Our work shows that there is room to substantially improve the performance of these clocks. As another example, sodium guidestars are important for imaging of space objects, and much of the early work on this important technology was done at the Starfire Optical Range at Kirtland Air Force Base with substantial input from the Principal Investigator of this grant.

Major Accomplishments

1. The book “Optically Pumped Atoms”

For several years, the principal investigator, W. Happer and two of his former graduate students, T.G. Walker and Y.-Y. Jau have been working on a book “Optically Pumped Atoms,” that summarizes some of the lessons learned from research supported by AFOSR through this grant and its predecessors [1]. The book came out in the spring of 2010, and the cover is shown in Fig. 1. This ready reference covers the most important facts about optical

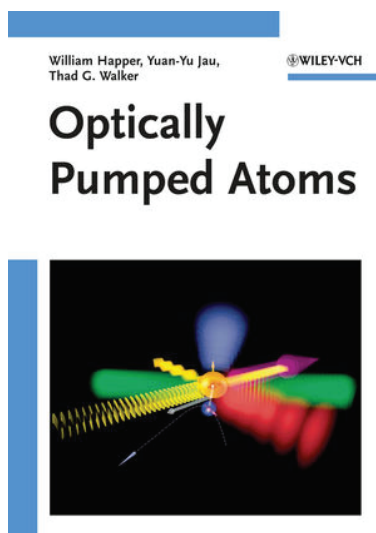


Figure 1: Book summarizing some of the research supported by this AFOSR grant.

pumping and spin relaxation of atoms. The authors show that systematic use of Liouville space, together with modern scientific computing software makes it practical to analyze the full, multilevel system of optically pumped atoms. Sections of MATLAB codes included in the text allow the reader to assemble quite sophisticated codes for modelling various optical-pumping phenomena.

2. Physics of gas-cell atomic clocks

The most widely used atomic clocks are based on the $0-0$ frequencies of Rb and Cs atoms. In these clocks, a Rb or Cs vapor is optically pumped by a lamp or a diode laser, and at the same time a hyperfine transition between ground-state sublevels is excited by magnetic resonance or by modulated light. In either case, it is customary to use a chemically inert buffer gas to keep the pumped atoms from diffusing too quickly to the walls of the resonance cell. As a result of collisions of the gas atoms or molecules with the optically-pumped alkali-metal atoms, the clock frequency is shifted from the ideal hyperfine frequency, the Bohr frequency for the “0-0” transition between the two ground-state sublevels with azimuthal quantum numbers $m = 0$. The resulting “pressure shifts,” together with the “light shifts”

of the clock frequency are one of the limits on clock accuracy. To get the best possible clock performance, it is necessary to thoroughly understand the pressure shifts. Practical clocks almost always contain a mixture of Ar and N₂ gas, with a mixing fraction that nearly cancels the temperature coefficient of the pressure shift. It has always been assumed that both gases cause a linear pressure shift, that is, if the partial pressure of the gas is doubled at the same temperature, the corresponding shift is doubled. Alkali-metal atoms are known to form loosely bound van der Waals molecules with Ar atoms, as sketched in Fig. 3. The molecules will also contribute to the shifts of the clock frequency, but the resulting shifts have substantial non-linear contributions at the low pressures that are used in practice.

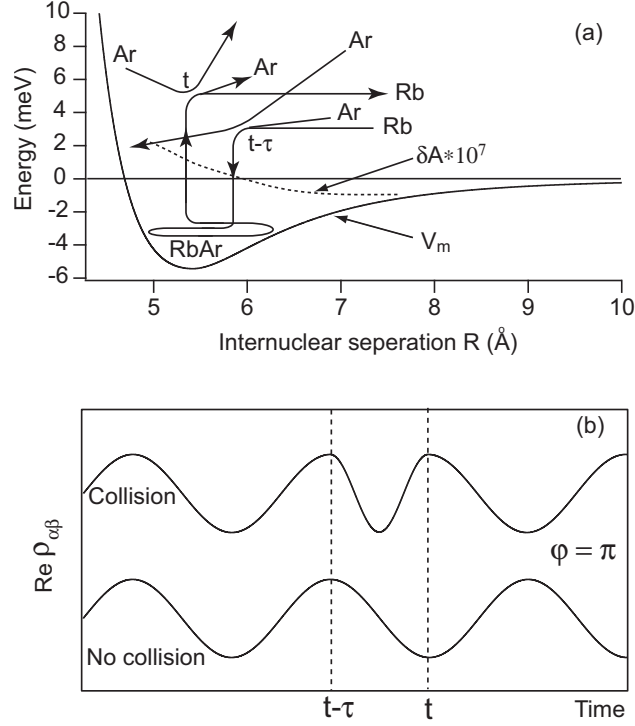


Figure 2: The top panel (a) shows the formation of a RbAr van der Waals molecule in the potential well V_m by a three-body collision, $\text{Rb} + \text{Ar} + \text{Ar} \rightarrow \text{RbAr} + \text{Ar}$, at the time $t - \tau$. The molecule is broken up in a second collision, $\text{RbAr} + \text{Ar} \rightarrow \text{Rb} + \text{Ar} + \text{Ar}$, at time t . The bottom panel (b) shows a (greatly-exaggerated) shift in the phase, ϕ , of the microwave coherence, $\rho_{\alpha\beta}$, for a bound atom due to the shift $\gamma \mathbf{N} \cdot \mathbf{S} + \delta A \mathbf{I} \cdot \mathbf{S}$ of the coupling of the electron spin \mathbf{S} with the nuclear spin \mathbf{I} while the Rb atom is part of a van der Waals molecule of rotational angular momentum \mathbf{N} .

Representative shifts measured with support from this grant are shown in Fig. 3. These non-linear shifts have been ignored in previous clock design, and when they are taken into account, there is a good chance that we can build simple, gas-cell clocks that can be primary standards, with no pressure shifts or light shifts, rather than secondary standards like all existing clocks.

In our continued studies of the hyperfine frequency shifts due to molecules, we have

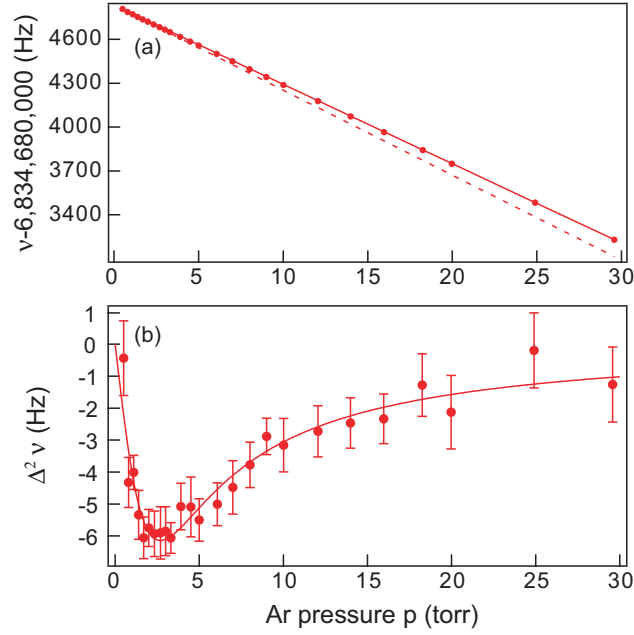


Figure 3: The points on the top panel (a) are measured 0–0 clock frequency of ^{87}Rb in Ar at a temperature of 40 C and a magnetic field $B = 2$ G. The error bars are too small to display. The solid line in (a) is the linear, limiting shift due to both binary collisions and to the formation and breakup of van der Waals molecules, as sketched in Fig. 2. The dashed line is the contribution from binary collisions alone. The points of the bottom panel (b) shows the molecular contribution, the difference between the two curves on the upper panel. More detail can be found in Ref. 2.

measured the pressure shifts of two additional important gases, neon and xenon. Neon is a common buffer gas in clocks based on coherent population trapping, where it is used as a non-quenching alternative to N_2 in gas mixtures with Ar that are optimized to reduce the temperature sensitivity of the clock. In our studies with neon, we discovered an important systematic error in our past measurements: our measurements are precise enough that unavoidable quadratic nonlinearities in common capacitance manometer pressure gauges are significant. We have been able to correct for this error, and so improve our measurements. Whether or not neon forms enough molecules to be detectable in gas cells has been a subject of debate in the past, and our improved measurements show an absence of molecule shifts to within experimental error.

Xenon, on the other hand, is known to form molecules in substantial numbers, and is a common gas in spin-exchange optical pumping. Our measurements show that molecules with xenon contributes a strikingly large shift that is opposite in sign to those with the similar gases Ar and Kr. From the sign discrepancy we have learned that the poorly-known hyperfine shift potential for Rb or Ce with Xe, which depends on the proximity to Xe, is qualitatively different from those with Ar and Kr. Additionally, the large size of the shifts with xenon has allowed us to resolve new, smaller features of the molecule shift which we think will lead to

improved understanding of the roles of different molecular vibration-rotation states in the molecule shifts.

Further work in progress is studying the dependence of molecule shifts on the isotope of Rb, which promises to provide new information about the poorly-known hyperfine shift potentials of Rb with Ar, Kr, and Xe.

The work on nonlinear pressure shifts is being led by Bart McGuyer, as part of his PhD research. During the course of his work he noticed that it is possible to substantially simplify the design of the feedback loops needed to: 1) lock the local oscillator of an atomic clock to the atomic hyperfine frequency and 2) to adjust the pump-laser frequency to the point of zero light shift. Normally, two lockin amplifiers are used to control the feedback loops for these two control loops, but Bart discovered that it is possible to use the in-phase and quadrature signals from a single lockin amplifier to accomplish the same goal. A comparison of the novel and traditional locking methods is shown in Fig. 4

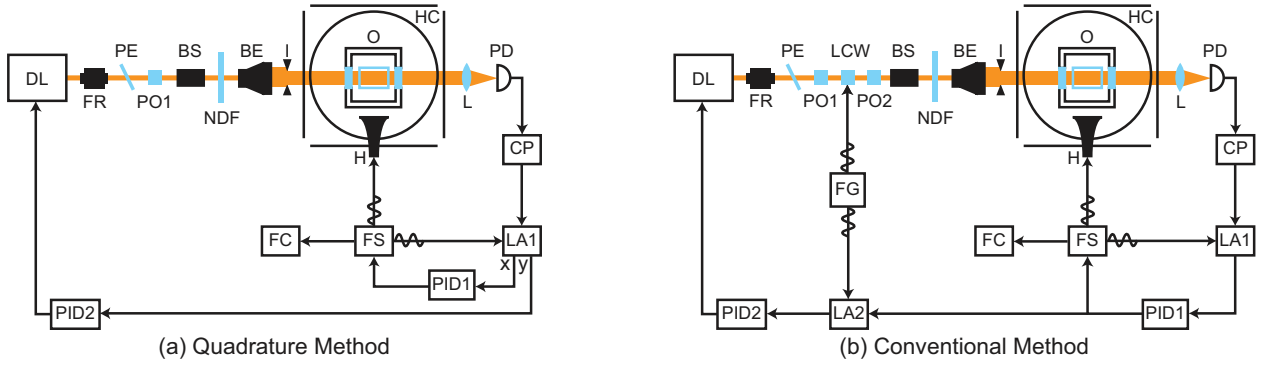


Figure 4: The new quadrature method for locking the crystal oscillator frequency and nulling the light shift. By using both the in-phase and quadrature signals from a single lock-in amplifier, one lockin amplifier can be eliminated. DL, diode laser; FR, Faraday rotator; PE, pellicle; PO, polarizer; LCW, liquid crystal wave; BS, beam shaper; NDF, neutral density filter; BE, beam expander; I, iris; O, oven; H, horn; HC, Helmholtz coils; L, lens; PD, photodetector; CP, current preamplifier; LA, lock-in amplifier; PID, PID controller; FS, frequency synthesizer; FC, frequency counter; and FG, function generator. More detail can be found in Ref. 4.

3. Optically-pumped metastable ^{129}Xe atoms.

Metastable noble-gas atoms are closely analogous to alkali-metal atoms in their ground state. Both atoms have a loosely-bound valence electron in an $ns_{1/2}$ orbital. For alkali-metal atoms, the non-valence electrons are in spherically symmetric, closed shells. For metastable noble-gas atoms, the non-valence not quite spherically symmetric, since there is a hole in the outermost $p_{3/2}$ shell, which is missing the $s_{1/2}$ valence electron. The spectrum of the atom is complicated by the interactions between the anisotropic core and the valence electron, but there remain many analogies to the spectrum of alkali-metal atoms.

In comparison to alkali-metal atoms, there are far fewer studies of metastable noble-gas atoms. Nevertheless, recent studies of cold atoms, atomic clocks, and atomic magnetometers, and hyperpolarized noble gas nuclei have stimulated the new interest in spin-polarized metastable noble-gas atoms. We have conducted a series of experiments to understand the physics of metastable xenon atoms in a sealed cell. We have completed studies of practical issues, like the lifetime of the discharged cell, as well as more fundamental issues like the broadening mechanisms of magnetic resonance linewidths. The major part of this work was carried out by Post Doctoral Research Assistant, Dr. Steven Morgan, and by Dr. Tian Xia, who included more details in his PhD Dissertation from Princeton University. More readily accessible details of this work can be found in Ref. 6.

4. Spin currents from optically pumped vapors to surfaces.

It has long been known that most cell walls for optically pumped atoms nearly completely destroy the spin of atoms that collide or adsorb on the walls. Most of the angular momentum lost by the impinging atoms probably goes into translational degrees of freedom of the wall material, but it is possible that some may go into the nuclear spins of the wall. Several years ago we showed that substantial amounts of nuclear spin can be deposited in the alkali-metal nuclei of hydride salt coatings on the walls of optical pumping cells [K. Ishikawa, B. Patton, Y.-Y. Jau and W. Happer, “Spin Transfer from an Optically Pumped Alkali Vapor to a Solid,” *Phys. Rev. Letters* **98**, 183004 (2007)]. A graduate student, Mr. Ben Olsen, has greatly improved the apparatus used in this initial work, and has also greatly improved our computer modelling capabilities, so that we now have a much better understanding of how the nuclear and electronic parts of the spin currents reach the cell walls. Mr. Olsen has also studied the use of high-field-forbidden transitions as especially effective ways to polarize nuclear spins at high magnetic fields.

5. Physics of sodium guidestar atoms.

Sodium guidestars are being ever more widely used for ground-based telescopes to correct for the atmospheric turbulence that limits the angular resolution of these devices. The idea, invented by Professor Happer in 1982, but heavily classified for many years, is to use a laser to excite the sodium atoms that occur naturally at an altitude of about 100 km above the earth’s surface. The backscattered light from the sodium atoms serves as an artificial star with which one can sample the turbulence of the atmosphere in the direction the laser points and correct for the distorting effects of turbulence with a deformable mirror. Because of the increasing use of sodium guidestars it seemed appropriate to look more carefully at the physics. In particular, the lasers used to produce sodium guidestars can optical pump of the sodium atoms into spin states that substantially enhance the scattering rate and improve the performance of the overall system, as illustrated in Fig. 5. The number density, $[N]$, of atmospheric molecules and atoms is so low at 100 km altitude, typically $[N] \approx 10^{14} \text{cm}^{-3}$, that the optical absorption lines are almost those of collision-free atoms. The homogeneous linewidth is so narrow that only the small fraction of atoms that happen to have the resonant Doppler shift can interact with a monochromatic pumping laser. Velocity-changing collisions allow spin-polarized atoms produced at the resonant velocity to populate the rest of velocity

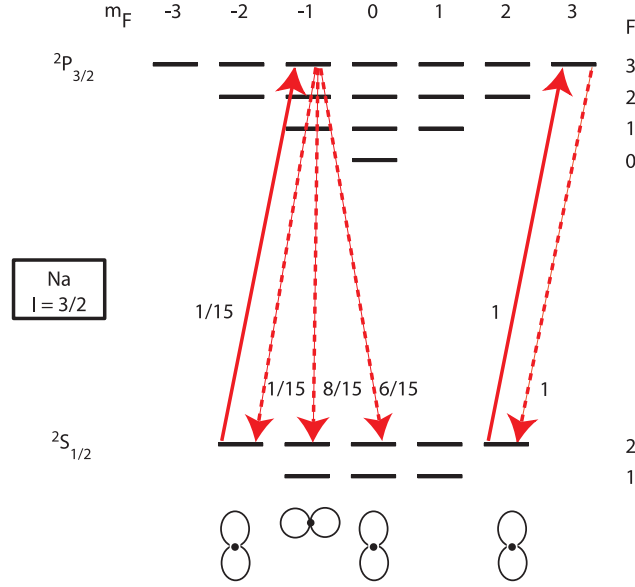


Figure 5: Substantial improvement of the return signal from sodium guidestars can be produced by optical pumping the atoms into “bright states,” for example, the sublevel with maximum azimuthal quantum number $m_F = 2$. As one can see from the sketch the fluorescence from this sublevel gives the maximum return to the ground, since it corresponds to a σ_+ transition with a radiation pattern proportional to $1 + \cos^2 \theta$, where θ is the direction of the emitted photon with respect to the direction of the laser beam, which defines the quantization axis of this figure. So the fluorescence back down the laser beam toward the adaptive optics receiver is a factor of 2 larger than for isotropic fluorescence. Other sublevels, for example, sublevel with $F = 2$ and $m_F = -2$ are less favorable, since they emit not only σ but also π radiation. π radiation cannot be observed at adaptive optics site on the ground because of its $\sin^2 \theta$ radiation pattern.

space. Spin precession around the geomagnetic field, with a magnitude $B \approx 1/2$ Gauss, is fast enough to substantially degrade any transverse spin polarization produced by the optical pumping in the time it takes for velocity-changing collisions to fill out the Maxwellian velocity distribution. Spin-flipping collisions with the “buffer gas” – the nitrogen molecules and the oxygen molecules and atoms of the residual atmosphere – can also degrade the spin polarization before an atom can be transferred from the pumped velocity group to other velocity groups. The relaxation and magnetic field conditions of sodium guidestars are almost never encountered in the laboratory, so initial experiments are being undertaken by graduate student, Natalie Kostinski, to investigate this physics for the first time.

The new mathematical tools that are needed to analyze sodium guidestar physics and related laboratory experiments turn out to have a long pedigree, going back to work by Rayleigh in 1891, and including important contributions by Einstein, Smoluchowski, Uhlenbeck and many others. In the “weak-collision” limit studied by Rayleigh, where the velocity changes per collision are small compared to the mean thermal velocity, the evolution of the velocity distributions is well described by Fokker-Planck equations for diffusion in a parabolic potential well. During the past year we have shown the solutions of these problems are facilitated by a new special function of the velocity, somewhat analogous to Bessel functions which are so useful for problems with cylindrical symmetry. We have developed efficient ways to generate and use these new special functions, and we have shown that there is a close relationship between the dissipative physics of weak velocity-changing collisions and the non-dissipative physics of the harmonic oscillator. This includes close analogs of the Glauber states of oscillators. The first publication of work in this area is Ref. 6.

Two undergraduate students, Ivana Dimitrovna, who graduated in 2010, and Bobby Marsland, who will graduate in 2011, have devoted senior theses to aspects of sodium guidestar physics. Two additional graduate students, Ben Olsen and Bart McGuyer, are also working on PhD thesis under Professor Happer’s guidance and financial support.

Personnel

AFOSR’s support for the training of students and postdoctoral research associates is one of the most important benefits of the grant to Princeton University and to the United States. The following personnel have received full or partial support from Grant AFOSR FA9550-07-1-0103:

Dr. William Happer, Principal Investigator.

Dr. Steven Morgan, Research Associate.

Mr. Xia Tian, Graduate Student.

Mr. Ben Olsen, Graduate Student.

Ms. Natalie Kostinski, Graduate Student.

Mr. Bart McGuyer, Graduate Student.

Ms. Ivana Dimitrovna, Undergraduate Student.

Mr. Bobby Marsland, Undergraduate Student.

Publications

Listed below are publications resulting from work published in the last few years with full or partial support from AFOSR.

1. T. G. Walker, Y.-Y. Jau and W. Happer *Optically Pumped Atoms*, Wiley-VCH GmbH Verlag, Weinheim (2010).
2. F. Gong, Y.-Y. Jau and W. Happer, Nonlinear Pressure Shifts of Alkali-Metal Atoms in Inert Gases, *Phys. Rev. Letters*, **100**, 233002 (2007).
3. J. Ma, A. Kishinevski, Y.-Y. Jau, C. Reuter and W. Happer, Modification of glass cell walls by rubidium vapor, *Phys. Rev. A*, **79**, 042905, (2009).
4. B. H. McGuyer, Y.-Y. Jau, and W. Happer Simple Method of Light-Shift Suppression in Optical Pumping Systems, *Applied Physics Letters*, **94**, 251110 (2009).
5. T. Xia, S. W. Morgan, Y.-Y. Jau and W. Happer, Polarization and Hyperfine Transitions of Metastable ^{129}Xe in Discharge Cells, *Phys. Rev. A*, **81**, 033419, (2010).
6. S. W. Morgan and W. Happer, Optically Pumped Atoms with Velocity- and Spin-Changing Collisions at Low Gas Pressure, *Phys. Rev. A*, **81**, 042703, (2010).

Interactions/Transitions

As an example of current Air Force research and development activities that have benefitted from the work of this AFOSR grant, the Principal Investigator recently received the following invitation:

“I am writing to ask if you would be available to present an invited talk at the Advanced High-Power Lasers meeting that will be held in Santa Fe (NM), June 6-10, 2011. The details of this meeting are described on the pages of the Directed Energy Professional Society web site (<http://www.deps.org/DEPSpages/AHPL11.html>). This year the meeting will include a symposium on high-power gas lasers. As you know, the goal of developing alkali vapor lasers that are optically pumped by diodes has been enthusiastically pursued in recent years. This has generated additional interest in the physics associated with optical pumping of alkali metals, line broadening processes and collisional energy transfer kinetics. Given your extensive and seminal contributions to this field, we would be most grateful for your participation, in the form of a 40 minute overview talk.”

1 New Inventions

Patents have been filed in connection with the quadrature clock locking, discussed in Ref. [4] and the pressure-shift-free gas-cell clock, based on the physics discussed in Ref. 2.

Honors/Awards

Professor Happer is a Fellow of the American Physical Society, the American Association for the Advancement of Science. He has been elected a member of the American Academy of Arts and Sciences, the National Academy of Sciences and the American Philosophical Society. He was awarded an Alfred P. Sloan Fellowship in 1966, an Alexander von Humboldt Award in 1976, the 1997 Broida Prize and the 1999 Davisson-Germer Prize of the American Physical Society, and the Thomas Alva Edison Patent Award in 2000.